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Orange opals from Buriti dos Montes, Piauí: solid inclusions as genetic guides

Opalas laranjas de Buriti dos Montes, Piauí: inclusões sólidas como guias genéticos

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Abstract

Orange opals from Buriti dos Montes (Piauí, northeastern Brazil) have gemological properties that favor their use as jewelry; these characteristics include their colors, transparency, relatively high stability and hardness. The exotic content of solid inclusions provides greater beauty to the opals of this region. These opals originated from hydrothermal processes and are found mainly as veinlets and veins in the sandstones of the Serra Grande Group, sectioned by diabase dikes and sills of the Sardinha Formation. Solid inclusions, such as bubbles, botryoidal aggregates, dendrites, and nodules, among others, consist mainly of kaolinite, hematite/goethite and quartz and influence the chemical composition of opals. Intense zoning of quartz crystals and high values of Ba and Fe suggest that opal deposits were formed in a hydrothermal environment. Diabase dykes could have been responsible for heating the hydrothermal fluids. Sandstones, rich in aqueous solutions, also contributed to the available silica for the saturation of these solutions, and fractures enabled the migration and entrapment of hydrothermal fluids, resulting in the mineralized veins.

Keywords: orange opal, solid inclusions, hydrothermal genesis.

Resumo

As opalas laranjas de Buriti dos Montes (Piauí, nordeste do Brasil) têm propriedades gemológicas que favorecem seu uso como jóias; essas características incluem as cores, transparência, dureza e estabilidade relativamente elevadas. O exótico conteúdo de inclusões sólidas proporciona maior beleza às opalas da região. Essas opalas foram originadas por processos hidrotermais e são encontradas, principalmente, em vênulas e veios nos arenitos do Grupo Serra Grande, seccionados por soleiras e diques de diabásio da Formação Sardinha. Inclusões sólidas, tais como bolhas, agregados botrioidais, dendritos e nódulos, entre outras, consistem, principalmente, de caulinita, hematita/goethita e quartzo e influenciam a composição química das opalas. O zoneamento intenso dos cristais de quartzo e os elevados valores de Ba e Fe sugerem que os depósitos de opala foram formados em ambiente hidrotermal. Os diques de diabásio teriam sido responsáveis pelo aquecimento dos fluidos hidrotermais. Os arenitos, ricos em soluções aquosas, também teriam contribuído com a sílica disponível para a saturação dessas soluções e as fraturas permitiram a migração e aprisionamento dos fluidos hidrotermais, resultando nos veios mineralizados.

Palavras-chave: *opala laranja, inclusões sólidas, gênese hidrotermal.*

1. Introduction

In the Piauí State of northeastern Brazil, Pedro II and Buriti dos Montes counties have the most important opal occurrences of the country. The gemological qualities of these opals are equivalent to the famous Australian opals. According to Gomes (2002), the orange opals from Buriti dos Montes do not exert the same fascination as the precious opals from Pedro II because they do not show a play-of-colors. However, this variety has been highly appreciated due to other features, which are also suitable for use in jewelry, such as their orange hues, transparency, hardness and relatively high stability. The exotic content of solid inclusions provides greater beauty to the orange opals found in this region. These inclusions occur isolated or in various arrangements with millimetric to centimetric size that are intrinsic to Buriti dos Montes opals. Similar solid inclusions have been described in Mexican opals by Koivula *et al.* (1983), Gübelin & Koivula (1986a, 1986b, 2005), Smith (1990) and Spencer *et al.* (1992); Mexican opals are also well known in the international gemological market. As shown by Gomes (2002), Gomes & Costa (2001), Marques (2011) and Marques *et al.* (2012), Piauí opals originated from hydrothermal processes. In this context, this paper discusses the morphology, mineralogical and chemical composition of solid inclusions, their interaction with the orange opals and their genetic significance.

Localization and access: Buriti dos Montes County is located in the northeastern part of the Piauí State, approximately 230 kilometers (km) east of the capital, Teresina. Access to Buriti dos Montes County from Teresina is facilitated by federal and state highways that intersect the region through Campo Maior and Castelo do Piauí.

Geological context and occurrence: The Piauí opals are hosted in the sedimentary rocks of the Parnaíba Paleozoic Basin, which involves three depositional cycles controlled by global tectonics, as proposed by Góes & Feijó (1994): the Silurian (Serra Grande Group), Devonian (Canindé Group) and Carboniferous-Triassic (Balsas Group) sequences. The Jurassic (Pastos Bons, Corda, Sardinha and Mosquito Formations) and Cretaceous (Grajaú, Codó and Itapecuru Formations, in the north, and Areado Group and Urucuia Formation, in the south) sequences were considered part of this basin but are directly related to the rupture of Gondwana; therefore, their evolutions are distinct from the Parnaíba Basin. Góes (1995) and Rossetti *et al.* (2001) suggested the names Alpercatas, Grajaú and Espigão-Mestre for these basins. The Serra Grande Group occurs within the limits of the Parnaíba Basin and presents itself in discordance with the overlying Canindé Group. According to Góes *et al.* (1990), the Serra Grande Group represents the first marine deposition in the Parnaíba Basin, corresponding to a complete transgressive-regressive cycle, where fluvial-deltaic to shallow marine sediments were deposited and consolidated into Ipu, Tianguá and Jaicós Formations. In general, continental sandstones and conglomerates with possible glacial influence (Ipu Formation) are present in gradual contacts with sandstones and marine shales (Tianguá Formation) that are overlapped by sandstones and fluvial conglomerates (Jaicós Formation). The Cretaceous basic intrusive magmatism occurs in the form of sills and dykes that constitute the Sardinha Formation and cross cut the Parnaíba Basin (Góes, 1995), including its base, where the rocks of Serra Grande Group that host the studied opals are found. These orange opals are found mainly as veins and veinlets, filling the fractures in Serra Grande Group sandstones. They also occur in cementing breccias in the contact zone between host rocks and mafic dykes. These primary deposits formed the source for the colluvial and paleochannel deposits. Locally, there are veins of quartz associated with opals with goethite dispersed between quartz crystals and opals (Gomes & Costa, 2001).

2. Materials and methods

One hundred forty six opal samples distributed into 20 lots, with an average of approximately 7 pieces each were used. These samples were collected by the geologist Érico Rodrigues Gomes. The opals and their inclusions were characterized by their morphology, micromorphology, and mineralogical and chemical compositions using the following analytical techniques:

X-Ray Powder Diffraction (XRD): XRD was carried out at the Mineral Characterization Laboratory of the Geosciences Institute of Federal University of Pará (IG-UFPA). XRD was used to determine the main mineral phases and crystalline order-disorder degree (crystallinity) of the opals, which were prepared by the powder method. We used an X-ray diffractometer (model X'Pert Pro MPD (PW 3040/60) PANalytical) with a PW3050/60 (θ-θ) goniometer and an X-ray tube ceramic with a Cu anode (Kα1 = 1.540598 Å) model PW3373/00, long fine focus, Ni Kβ filter, X'Celerator Real Time Multiple Scanning (RTMS) detector in scanning mode with an active length of 2.122º. Scans were conducted from 5° to 75° 2θ with a voltage of 40 kV, a 30 mA current, a 0.02° step in 2θ and 10 s/step, an automatic and 4° anti-scattering slit, a 10-mm mask, and a spinning motion of sample with 1 rps. The data were processed and interpreted using an X'Pert Data Collector and X'Pert High Score software, both from PANalytical.

Inductively Coupled Plasma-Mass Spectrometry (ICP-MS): ICP-MS was conducted at Acme Analytical Laboratories Ltd. to determine the bulk chemical composition of the opals. The samples were previously powdered into aliquots of 200 mg, melted with lithium metaborate and tetraborate, and then dissolved with nitric acid. The major C, S, and trace elements, including rare earth elements (REE), were determined.

The loss on ignition (LOI) is given by the difference in weight after calcination at 1000°C.

Optical stereomicroscopy: All samples were observed with a Zeiss stereomicroscope (model Stemi 2000-C), described according to their macroscopic features and photographed.

Scanning Electron Microscopy (SEM/EDS/CL): Fragments and polished thin sections of opals, emphasizing their inclusions, were used for the morphological and mineralogical study using secondary electron (SE) images and energy dispersive spectrometry (EDS) semi-quantitative chemical analysis. All fragments of samples were fixed on stubs with carbon double-sided tape and coated with gold for 2 minutes. The polished thin sections were also used to obtain backscattered (BSE) and cathodoluminescence (CL) images, in addition to EDS analysis. These thin sections were coated with gold for 2 minutes for EDS analysis and 30 seconds for CL images, with a coating thickness of 15 nm and 3 nm, respectively. A LEO ZEISS SEM (model 1430) with a Sirius-Gresham EDS detector and a Gatan Mono-CL belonging to the Laboratory of Scanning Electron Microscopy of the UFPA were used.

Operating conditions for the SE and BSE images and the EDS analysis were as follows: beam current = 90 µa, accelerating voltage = 20 kV, work distance = 15 mm, scanning time = 30 s with 3000 to 4000 cycles per second (c/s) for each analysis. The conditions for the CL images were the same but with work distance = 13 mm.

Opal Structure

The opals from Buriti dos Montes are semitransparent to translucent and have a wide range of hues, from light yellow to brownish red. In the SEM images, it is possible to see that the interstices between silica spheres forming the opal were partially filled by opal cement, masking the typical subspherulitic aspect, giving rise to an almost massive feature (Figure 1A, B). The sphere diameters range from 150 to 500 nm. Irregular spheres and compact arrangements do not allow for the production of a play-of-colors, an effect that manifests itself only when the spheres are perfect, uniform in size and have a regular packing without any interstices filling (Wollaert *et al.*, 1990). However, these opals have good transparency, which may be related to the size of the spaces between the spheres. According to Schwartz (1984), small interstices provide greater transparency to opals.

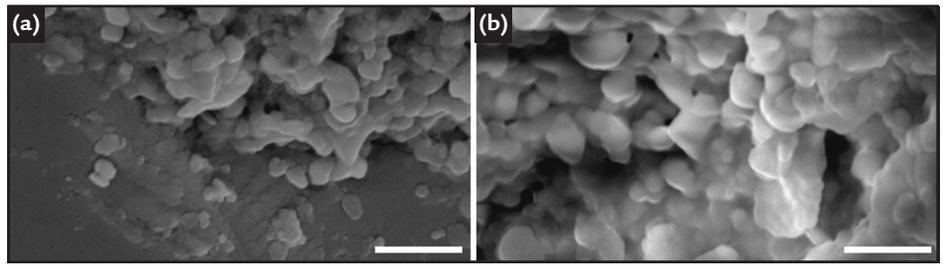

Figure 1
A, B - Subspherulitic arrangement of the orange opals, whose cementation generates a massive aspect. Secondary electron images obtained using SEM. Scale bar = 1.5 µm.

Solid Inclusions Morphology

The different types of solid inclusions in the studied opals were identified and classified according to morphology using criteria such as shape, color, size and texture as well as the three-dimensional arrangement produced within the opals.

The inclusions, named bubbles, correspond to rounded cavities with diameters between 5 and 20 mm and are cream-white or brownish according to the filling, usually of host sandstones. Some bubbles have a white film on the inner wall of the cavity, while others are completely empty, which most likely indicates already-released gas content. There are bubbles surrounded by tension fractures (Figure 2A) with an external appearance of microconcretions and filled with opaline silica (Figure 2B). The botryoidal inclusions are sandy aggregates of the host rock, with a botryoidal aspect and a general tendency towards rounding. Their color varies from white to cream and brown to reddish tones. The spatial arrangement of these inclusions within the opals allows for characterizing different morphologies, such as coral reefs, stalactites, stalagmites and columns (Figure 2C). Dendritic inclusions (Figure 2D), with a typical arborescent aspect, are dark, usually brown, and are associated with botryoidal inclusions, bubbles and fractures.

The tabular or lamellar inclusions (Figure 2E) are groups of parallel lamellae or platelets of sandy material. Microcrystals, isolated or grouped, are euhedral and well formed. Microcrystals

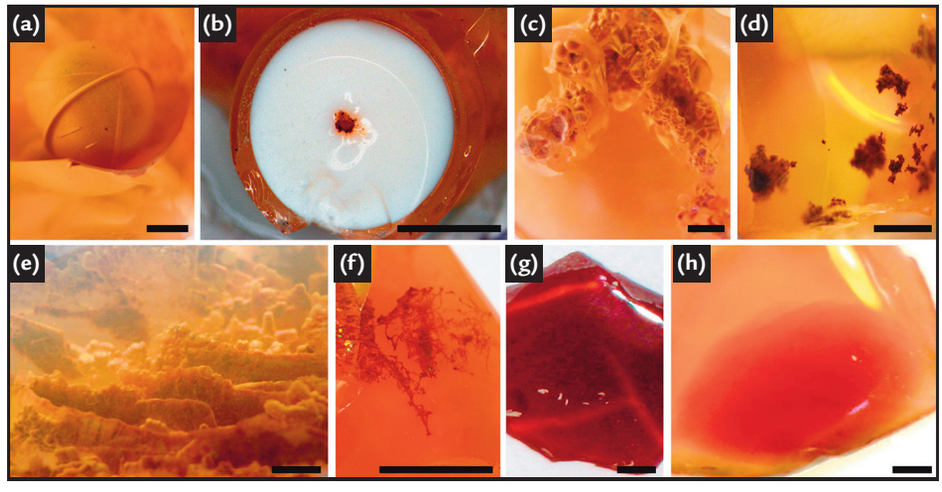

Figure 2
Opals from Buriti dos Montes with the main solid inclusions and associated features. A - bubble with tension fracture, B - white opal bubble with Fe oxy-hydroxides in the center, C - botryoidal-columnar inclusion with tension fractures, D - dendrites, E - lamellar inclusions with botryoidal terminations, F - helicitic inclusions with dry fractures, G - sealed fractures in red opal, H - red patch in orange opal. Scale bar = 1 mm

of quartz inclusions in these opals are frequent. Nodules, composed of materials from the host rocks, represent sandy fragments that were not completely dissolved during the migration of hydrothermal fluids. The helicitic inclusions or cobwebs (Figure 2F) are formed by brown, irregular tiny needles, similar to strands of hair, together constituting an arrangement similar to a cobweb. The tubes are cylindrical and elongated channels, sometimes curved, with millimetric diameter, which may be empty or partially or completely filled with sandy material. The filling gives reddish or whitish colors to the tubes.

Some features, such as flow structures (striations that represent the probable migration of fluids through fractured host sandstone) and fractures, occur in isolation or in association with botryoidal inclusions and bubbles. There are sealed fractures (Figure 2G - totally filled by opal), dry fractures (unfilled fractures), tension fractures (semicircular fractures that surround other inclusions), superficial fractures (external fractures, non-penetrative) and cracking fractures (group of interconnected fractures that delimit small polygons with penetrative character). Patches and color zones (Figure 2H), also frequent, are portions of opal with variations of the main color.

Mineralogical Caracterization

The mineralogical analysis by XRD revealed different order-disorder degrees in orange opals, from amorphous opals (opal-A) to cristobalite-tridymite opals (opal-CT). Samples GB 7-28, GB 8-37 and GB 6-2 are typical of opal-A, whose diffraction patterns show a main peak with d = 4.11 Å (4.10/4.12), broad and high background, and another incipient in d = 2.50/2.51 Å. The high background and low resolution of the peak d = 4.32 Å, sometimes almost imperceptible, suggest low-order crystalline, approaching an amorphous state. Samples GB 2-14 and GB 9-6 are typically opal-CT, characterized by interplane distances d = 4.10 Å (4.11), 4.31 Å (4.32/4.33), 2.50 Å, 2.05 Å and 1.63 Å. Among the terms defined as amorphous and cristobalite-tridymite, there are intermediate stages of order-disorder crystalline, as in the GB 1-24, GB 5-5, GB 4-5, GB 3-20 and GB 10 samples (Figure 3A, B). The presence of kaolinite in the solid inclusions is indicated by reflections d = 7.17 Å (7.18/7.19) and 3.58 Å. The interplane spacings d = 3.34 Å (3.35), 1.54 Å and 1.44 Å refer to quartz microcrystal inclusions in the opals. These quartz microcrystals as well as botryoidal and lamellar inclusions, nodules and tubes formed predominantly by kaolinite have been confirmed by EDS analysis.

In addition to these minerals, hematite and/or goethite were also found in the darker portions of the botryoidal, dendritic and helicitic inclusions as well as in the nodules and in the center of a bubble formed by white opal within yellow opal. Opaline pseudomorphs of gypsum or barite (Figure 4A, B) were found locally, filling small cavities in opals. EDS analysis indicated only Si and O, showing that these inclusions were completely replaced by the opaline silica, preserving, however, the shape of the crystals. Hyaline quartz also occurs in small druses and accompanying veins of opal. When imaged by CL using SEM, these crystals reveal a pattern of concentric zoning perpendicular to the c-axis, showing various intensities of luminescence. This feature is diagnostic of hydrothermal origin (Rusk et al., 2006; 2008) and contrasts strongly with the extremely homogeneous texture, i.e., without zoning, exhibited by grains of detrital quartz of the sandstones that host the mineralization. In CL images, venules of hydrothermal quartz overgrown on detrital quartz were observed (Figure 5A, B), indicating that fluids migrated through the interstices of the grains into the sandstones. The thickness of hydrothermal quartz veinlets gradually increases toward the ore veins, until the detrital grains are completely consumed, leaving neoformed euhedral quartz crystals (Figure 5C).

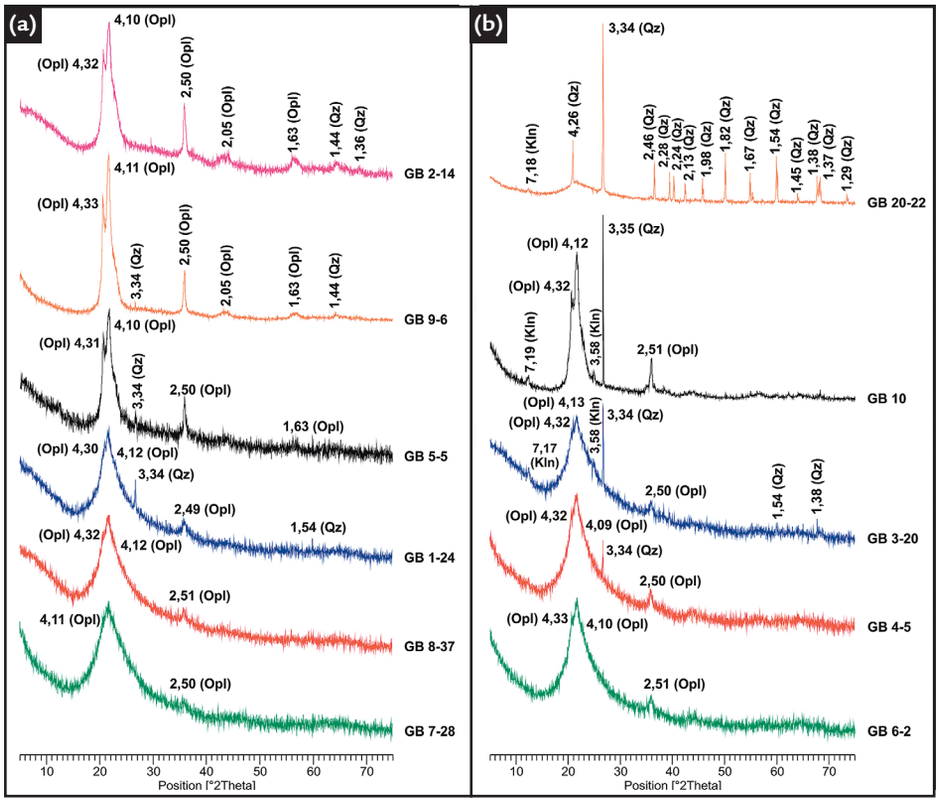

Figure 3
A, B - X-ray diffraction diagrams showing the different order-disorder degrees of the studied opals, with variation between opal-A and CT, and intermediate levels, from the base upwards. Kln - kaolinite, Opl - opal, Qz - quartz (abbreviations of mineral names based on Whitney & Evans, 2010).

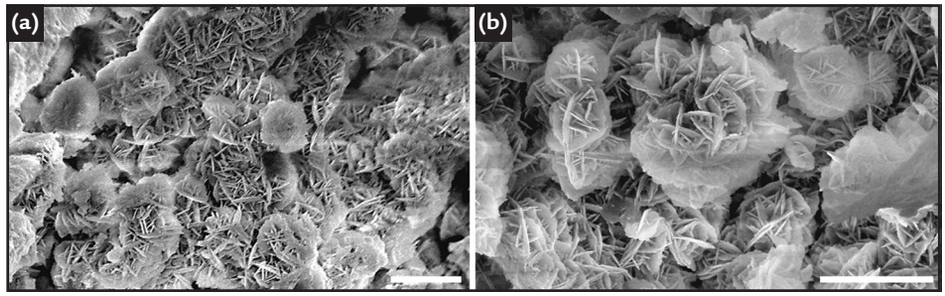

Figure 4
A, B - Pseudomorphs of gypsum or barite included in orange opals. Secondary electron images obtained using SEM.

Scale bar = 5 µm.

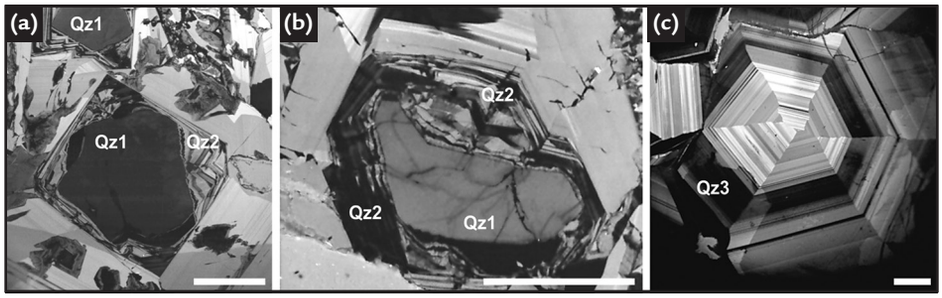

Figure 5
A, B - Detrital quartz grains (Qz1) of sandstones that host the opal mineralization with hydrothermal quartz overgrowth (Qz2). C - Euhedral quartz crystal (Qz3) with internal concentric zoning. Cathodoluminescence images obtained using SEM.

Scale bar = 100 µm. Qz - quartz (abbreviations of mineral names based on Whitney & Evans, 2010).

Chemical Composition

The opals from Buriti dos Montes are composed mainly of SiO_2 (90.14 % on average) and 8.03 % water (based on LOI), 1.32 % Al_2O_3 and 0.2 % Fe_2O_3. The SiO_2 contents correlate negatively with Al_2O_3 and Fe_2O_3, most likely due to the mineral inclusions, such as kaolinite and hematite. Part of the silica-contents is also found as quartz inclusions. Al can also be a potential Si replacement in the opal structures. Among the analyzed trace elements, only the high values of Ba are highlighted, ranging between 195 and 1373 parts per million (ppm), with an average of 808 ppm (Figure 6A). Other trace-element contents are very close to the detection limits. Gomes (2002) emphasizes that barite veins are common in the sandstones that host opals. The GB 4 sample, which is richer in Al_2O_3 and Fe_2O_3, displays the highest contents of REEs (Figure 6B) and trace elements, most likely due to an association with the solid inclusions of hematite and kaolinite. From EDS analysis, it was possible to identify higher contents of Fe in patches and dark colored zones than in the opal matrix. Similarly, reaction borders surrounding helicitic and dendritic inclusions have higher contents of Fe, with an average of 4500 ppm compared with the surrounding opal with an average of 1700 ppm of Fe.

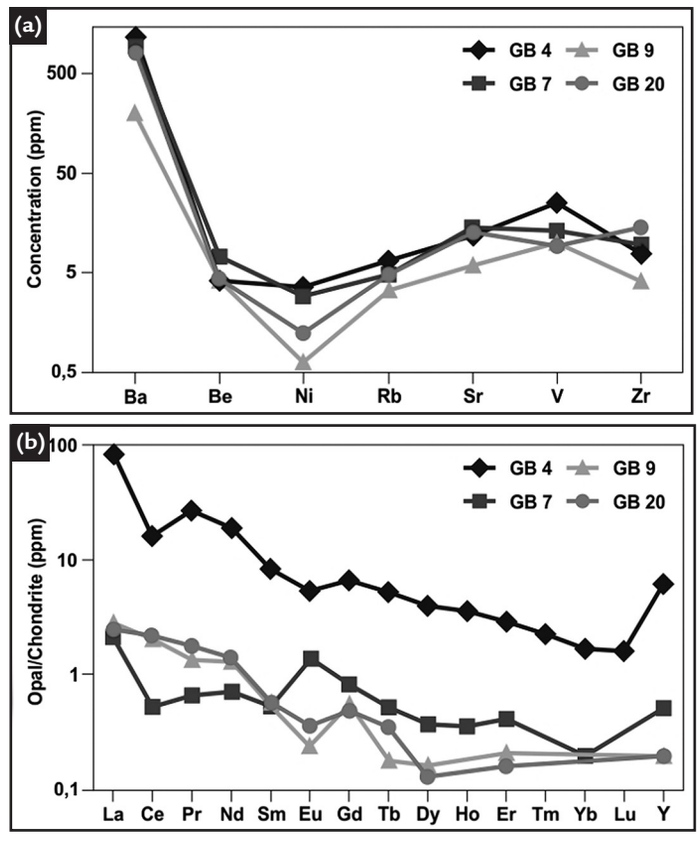

Figure 6
A - Multi-elements diagram with the distribution of trace elements in the opals. Trace element concentrations (ppm) are relatively higher in sample GB 4. B - Diagram showing the distribution of REE in these opals and considerable enrichment in sample GB 4. The contents were normalized by chondrite of Taylor & McLennan (1985).

3. Discussion and conclusions

The vast content of solid inclusions, their arrangements, contact relationships, mineralogy and chemical composition reinforce the hydrothermal genesis of orange opals from Buriti dos Montes as suggested by Gomes (2002), and discussed by Marques (2011). Most inclusions are formed by materials from the host rocks or simply from particles that were involved by fluids during their migration through the fractures in these sandstones. The hydrothermal fluid flow is evident in flow structures present in the opals and on features representing partial dissolution of some inclusions (e.g., the reaction borders around the helicitic and dendritic inclusions). Other features, such as sealed fractures, patches and zoning of colors, refer to late fluid flow and partial dissolution of hematitic plasma particles. The color zoning was created by neoformation of Fe oxy-hydroxides such as hematite and goethite within the opals. The possible existence of micro and nano-inclusions, rich in these minerals can also contribute to this effect (Fritsch et al., 1999; 2002; 2006; Steffen, 2000; Gaillou et al., 2008). The lamellar inclusions reveal a structure that can originate from the deformation of sandstones assimilated by fluids, considering that the mineralized area is under the influence of the Transbrasiliano Lineament. The opals from Buriti dos Montes are marked by high contents of Ba and Fe. Ba may possibly constitute inclusions of barite, a common mineral in rocks of the region. Fe occurs as oxy-hydroxides in solid inclusions, which give the distinct orange color to these opals. The presence of barite, hematite and kaolinite also reinforces the context of a hydrothermal environment for the origin of these opals. This hydrothermal genesis is also confirmed by the presence of hyaline quartz with intense zoning in CL, in lateral contact with opal in the veins, and as microcrystals included in the opals. This hypothesis is corroborated by the morphology of the inclusions, by the mineral assemblage characterized by hematite, barite and kaolinite, and by the simultaneous occurrence of inclusions and veins of typically hydrothermal quartz.

This hydrothermal environment was most likely conditioned by the diabases of the Sardinha Formation, which acted as the heat source responsible for heating the aqueous solutions present in the sandstones of the Serra Grande Group. These sandstones also contributed to most of the solubilized silica necessary for the saturation of the solution, while extensive and numerous fractures were indispensable for trapping these hydrothermal solutions, thereby hosting the mineralized veins.

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